

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents
United States Patent and Trademark
Office
Box PCT
Washington, D.C. 20231
ÉTATS-UNIS D'AMÉRIQUE

in its capacity as elected Office

Date of mailing:

24 February 2000 (24.02.00)

International application No.:

PCT/EP99/05577

Applicant's or agent's file reference:

75858

International filing date:

03 August 1999 (03.08.99)

Priority date:

15 August 1998 (15.08.98)

Applicant:

ZANIBELLI, Laura et al

1. The designated Office is hereby notified of its election made:



in the demand filed with the International preliminary Examining Authority on:

04 January 2000 (04.01.00)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer:

J. Zahra

Telephone No.: (41-22) 338.83.38

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

| | | |
|---|---|--|
| Applicant's or agent's file reference 75858 | FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below. | |
| International application No. PCT/EP 99/05577 | International filing date (day/month/year) 03/08/1999 | (Earliest) Priority Date (day/month/year) 15/08/1998 |
| Applicant ENITECNOLOGIE, S.P.A. et al. | | |

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 6 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☒ Unity of invention is lacking (see Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawings to be published with the abstract is Figure No.

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

1
☐ None of the figures.

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1a,c-29,31-37,42-45

Catalyst containing FER type zeolite

2. Claims: 1b,30,38-41

Catalyst containing phosphorous

INTERNATIONAL SEARCH REPORT

International Application No

/EP 99/05577

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C10G45/12 C10G45/64 C10G45/08 B01J23/85 B01J29/69
 B01J27/185 B01J27/188

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C10G B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|---|---|
| X | US 5 378 352 A (DEGNAN THOMAS F ET AL) 3 January 1995 (1995-01-03) cited in the application | 1,3-15, 42 |
| A | claims 1-3,6-10,14-16 column 5, line 1 - line 14 column 5, line 43 -column 6, line 10 column 6, line 32 - line 34 table 1 | 32,43 |
| X | US 5 576 256 A (GARCIA WOLFGANG ET AL) 19 November 1996 (1996-11-19) cited in the application | 1-8,10, 12,13, 18,29, 31,38-45 |
| | column 2, line 64 -column 3, line 23 column 5, line 57 -column 6, line 43 examples 2,4-7 | |
| | --- -/-- | |

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents :

A document defining the general state of the art which is not considered to be of particular relevance

E earlier document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

& document member of the same patent family

Date of the actual completion of the international search

10 November 1999

Date of mailing of the international search report

24. 11. 99

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
 NL - 2280 HV Rijswijk
 Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
 Fax: (+31-70) 340-3016

Authorized officer

Zuurdeeg, B

INTERNATIONAL SEARCH REPORT

International Application No

T/EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|--|
| X A | US 4 629 717 A (CHAO TAI-HSIANG) 16 December 1986 (1986-12-16) claims 1,4,5,10,11,15 column 7, line 64 -column 8, line 35 column 9, line 8 - line 21 example VI --- | 1,38-41 1,5-13, 29-31, 33,36,37 |
| X | EP 0 448 117 A (COSMO SOGO KENKYUSHO KK ;COSMO OIL CO LTD (JP)) 25 September 1991 (1991-09-25) claims 1-3,6,7,9,18 page 6, line 29 - line 35 page 7, line 54 -page 8, line 1 page 8, line 17 - line 21 example 8 --- | 1,38-41 |
| A | US 5 482 617 A (COLLINS NICK A ET AL) 9 January 1996 (1996-01-09) claims 1-6,8-10,13-16,18-25 column 5, line 21 - line 42 --- | 1,3-5, 12-16, 25,31, 43,45 |
| A | EP 0 449 144 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 October 1991 (1991-10-02) claims 1,3,8,10,11,17-19-22,24 page 5, line 20 -page 6, line 38 page 9, line 34 - line 44 tables 1-3,5 --- | 1,3-15, 31,32 |
| A | EP 0 665 280 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 August 1995 (1995-08-02) the whole document --- | 1-8,12, 13,33,37 |
| A | US 4 808 560 A (OLECK STEPHEN M) 28 February 1989 (1989-02-28) claims 1,2,4,7,8 column 6, line 23 - line 68 --- | 1,3-16 |
| A | EP 0 442 159 A (SHELL INT RESEARCH) 21 August 1991 (1991-08-21) cited in the application claims 1-9 page 5, line 55 -page 6, line 17 --- | 1,25,32 |
| | --- -/-- | |

INTERNATIONAL SEARCH REPORT

International Application No

EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category ° | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|-----------------------|
| A | <p> DATABASE WPI Section Ch, Week 8630 Derwent Publications Ltd., London, GB; Class H04, AN 86-193476 XP002089923 & JP 61 126196 A (JUSHITSUYU TAISAKU GIJUTSU), 13 June 1986 (1986-06-13) abstract ----- </p> | |

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

T/EP 99/05577

| Patent document cited in search report | | Publication date | Patent family member(s) | Publication date |
|---|---|---------------------|---|--|
| US 5378352 | A | 03-01-1995 | AU 658937 B AU 2734092 A CA 2081758 A DE 69222396 D DE 69222396 T EP 0543529 A ES 2106833 T JP 5247474 A | 04-05-1995 20-05-1993 20-05-1993 30-10-1997 26-02-1998 26-05-1993 16-11-1997 24-09-1993 |
| US 5576256 | A | 19-11-1996 | BR 9502505 A DE 19518607 A FR 2720073 A GB 2289689 A,B GB 2323094 A,B NL 1000428 C NL 1000428 A US 5591324 A US 5770047 A | 27-02-1996 30-11-1995 24-11-1995 29-11-1995 16-09-1998 24-12-1996 23-11-1995 07-01-1997 23-06-1998 |
| US 4629717 | A | 16-12-1986 | US 4727209 A | 23-02-1988 |
| EP 0448117 | A | 25-09-1991 | JP 2789489 B JP 3275142 A JP 4007044 A DE 69104247 D DE 69104247 T US 5182250 A US 5244858 A | 20-08-1998 05-12-1991 10-01-1992 03-11-1994 09-03-1995 26-01-1993 14-09-1993 |
| US 5482617 | A | 09-01-1996 | NONE | |
| EP 0449144 | A | 02-10-1991 | JP 2547115 B JP 3284354 A DE 69103058 D US 5187133 A | 23-10-1996 16-12-1991 01-09-1994 16-02-1993 |
| EP 0665280 | A | 02-08-1995 | JP 7197038 A JP 2875148 B JP 7194984 A | 01-08-1995 24-03-1999 01-08-1995 |
| US 4808560 | A | 28-02-1989 | NONE | |
| EP 0442159 | A | 21-08-1991 | US 5043520 A DE 69009727 D DE 69009727 T | 27-08-1991 14-07-1994 22-09-1994 |
| JP 61126196 | A | 13-06-1986 | JP 1902513 C JP 6031333 B | 08-02-1995 27-04-1994 |

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: Laura ZANIBELLI, et al

SERIAL NUMBER: NEW U.S. PCT APPLICATION (based on PCT/EP99/05577)

FILED: HERewith

**FOR: PROCESS AND CATALYSTS FOR UPGRADING OF HYDROCARBONS
BOILING IN THE NAPHTHA RANGE****REQUEST FOR CONSIDERATION OF DOCUMENTS
CITED IN INTERNATIONAL SEARCH REPORT**Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

In the matter of the above-identified application for patent, notice is hereby given that applicant(s) request that the Examiner consider the documents cited in the International Search Report according to MPEP §609 and so indicate by a statement in the first Office Action that the information has been considered. When the Form PCT/DO/EO/903 indicates both the search report and copies of the documents are present in the national stage file, there is no requirement for the applicant(s) to submit them (1156 O.G. 91 November 23, 1993).

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.

Norman F. Oblon
Attorney of Record
Registration No. 24,618
Surinder Sachar
Attorney of Record
Registration No. 34,423**22850**(703) 413-3000
Fax No. (703) 413-2220
(OSMMN 1/97)

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/EP 99/05577

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C10G45/12 C10G45/64 C10G45/08 B01J23/85 B01J29/69
B01J27/185 B01J27/188

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C10G B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|---|---|
| X | US 5 378 352 A (DEGNAN THOMAS F ET AL) 3 January 1995 (1995-01-03) cited in the application | 1,3-15, 42 |
| A | claims 1-3,6-10,14-16 column 5, line 1 - line 14 column 5, line 43 -column 6, line 10 column 6, line 32 - line 34 table 1 | 32,43 |
| X | US 5 576 256 A (GARCIA WOLFGANG ET AL) 19 November 1996 (1996-11-19) cited in the application | 1-8,10, 12,13, 18,29, 31,38-45 |
| | column 2, line 64 -column 3, line 23 column 5, line 57 -column 6, line 43 examples 2,4-7 | |



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *Z* document member of the same patent family

Date of the actual completion of the international search

10 November 1999

Date of mailing of the international search report

24.11.99

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Zuurdeeg, B

INTERNATIONAL SEARCH REPORT

Int. Application No
PCT/EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|--|
| X A | US 4 629 717 A (CHAO TAI-HSIANG) 16 December 1986 (1986-12-16) claims 1,4,5,10,11,15 column 7, line 64 -column 8, line 35 column 9, line 8 - line 21 example VI --- | 1,38-41 1,5-13, 29-31, 33,36,37 |
| X | EP 0 448 117 A (COSMO SOGO KENKYUSHO KK ;COSMO OIL CO LTD (JP)) 25 September 1991 (1991-09-25) claims 1-3,6,7,9,18 page 6, line 29 - line 35 page 7, line 54 -page 8, line 1 page 8, line 17 - line 21 example 8 --- | 1,38-41 |
| A | US 5 482 617 A (COLLINS NICK A ET AL) 9 January 1996 (1996-01-09) claims 1-6,8-10,13-16,18-25 column 5, line 21 - line 42 --- | 1,3-5, 12-16, 25,31, 43,45 |
| A | EP 0 449 144 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 October 1991 (1991-10-02) claims 1,3,8,10,11,17-19-22,24 page 5, line 20 -page 6, line 38 page 9, line 34 - line 44 tables 1-3,5 --- | 1,3-15, 31,32 |
| A | EP 0 665 280 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 August 1995 (1995-08-02) the whole document --- | 1-8,12, 13,33,37 |
| A | US 4 808 560 A (OLECK STEPHEN M) 28 February 1989 (1989-02-28) claims 1,2,4,7,8 column 6, line 23 - line 68 --- | 1,3-16 |
| A | EP 0 442 159 A (SHELL INT RESEARCH) 21 August 1991 (1991-08-21) cited in the application claims 1-9 page 5, line 55 -page 6, line 17 --- | 1,25,32 |
| | -/-- | |

INTERNATIONAL SEARCH REPORT

In. International Application No

PCT/EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|-----------------------|
| A | <p> DATABASE WPI Section Ch, Week 8630 Derwent Publications Ltd., London, GB; Class H04, AN 86-193476 XP002089923 & JP 61 126196 A (JUSHITSUYU TAISAKU GIJUTSU), 13 June 1986 (1986-06-13) abstract ----- </p> | |

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP 99/05577

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/EP 99/05577

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1a,c-29,31-37,42-45

Catalyst containing FER type zeolite

2. Claims: 1b,30,38-41

Catalyst containing phosphorous

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/05577

| Patent document cited in search report | Publication date | Patent family member(s) | Publication date |
|---|---------------------|----------------------------|---------------------|
| US 5378352 A | 03-01-1995 | AU 658937 B | 04-05-1995 |
| | | AU 2734092 A | 20-05-1993 |
| | | CA 2081758 A | 20-05-1993 |
| | | DE 69222396 D | 30-10-1997 |
| | | DE 69222396 T | 26-02-1998 |
| | | EP 0543529 A | 26-05-1993 |
| | | ES 2106833 T | 16-11-1997 |
| | | JP 5247474 A | 24-09-1993 |
| US 5576256 A | 19-11-1996 | BR 9502505 A | 27-02-1996 |
| | | DE 19518607 A | 30-11-1995 |
| | | FR 2720073 A | 24-11-1995 |
| | | GB 2289689 A,B | 29-11-1995 |
| | | GB 2323094 A,B | 16-09-1998 |
| | | NL 1000428 C | 24-12-1996 |
| | | NL 1000428 A | 23-11-1995 |
| | | US 5591324 A | 07-01-1997 |
| US 4629717 A | 16-12-1986 | US 4727209 A | 23-02-1988 |
| | | | |
| EP 0448117 A | 25-09-1991 | JP 2789489 B | 20-08-1998 |
| | | JP 3275142 A | 05-12-1991 |
| | | JP 4007044 A | 10-01-1992 |
| | | DE 69104247 D | 03-11-1994 |
| | | DE 69104247 T | 09-03-1995 |
| | | US 5182250 A | 26-01-1993 |
| | | US 5244858 A | 14-09-1993 |
| US 5482617 A | 09-01-1996 | NONE | |
| EP 0449144 A | 02-10-1991 | JP 2547115 B | 23-10-1996 |
| | | JP 3284354 A | 16-12-1991 |
| | | DE 69103058 D | 01-09-1994 |
| | | US 5187133 A | 16-02-1993 |
| EP 0665280 A | 02-08-1995 | JP 7197038 A | 01-08-1995 |
| | | JP 2875148 B | 24-03-1999 |
| | | JP 7194984 A | 01-08-1995 |
| US 4808560 A | 28-02-1989 | NONE | |
| EP 0442159 A | 21-08-1991 | US 5043520 A | 27-08-1991 |
| | | DE 69009727 D | 14-07-1994 |
| | | DE 69009727 T | 22-09-1994 |
| JP 61126196 A | 13-06-1986 | JP 1902513 C | 08-02-1995 |
| | | JP 6031333 B | 27-04-1994 |

PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

De Gregori, A.
ING. BARZANO' & ZANARDO
MILANO S.P.A.
Via Borgonuovo, 10
20121 Milano
ITALIE

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT (PCT Rule 71.1)

Date of mailing
(day/month/year) 04.12.2000

Applicant's or agent's file reference
Cal 75859

IMPORTANT NOTIFICATION

International application No.
PCT/EP99/05577

International filing date (day/month/year)
03/08/1999

Priority date (day/month/year)
15/08/1998

Applicant
ENITECNOLOGIE, S.P.A. et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Ing. B. & Z. MILANO
06. DIC. 2000

Name and mailing address of the IPEA/



European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0 Tx: 523656 epmu d
Fax: +49 89 2399 - 4465

Authorized officer

Gregoire, J-P

Tel. +49 89 2399-8041



INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

| | | | |
|---|--|--|---|
| Applicant's or agent's file reference Cal 75859 | FOR FURTHER ACTION | | See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416) |
| International application No. PCT/EP99/05577 | International filing date (day/month/year) 03/08/1999 | Priority date (day/month/year) 15/08/1998 | |
| International Patent Classification (IPC) or national classification and IPC C10G45/12 | | | |
| Applicant ENITECNOLOGIE, S.P.A. et al. | | | |

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 12 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

| | |
|---|--|
| Date of submission of the demand 04/01/2000 | Date of completion of this report 04.12.2000 |
| Name and mailing address of the international preliminary examining authority: <div style="display: flex; align-items: center;"> <div> European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465 </div> </div> | Authorized officer de Cauwer, R Telephone No. +49 89 2399 7344 |



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/05577

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:

Description, pages:

1-52 as originally filed

Claims, No.:

1-29 as originally filed

30-46 as received on 06/11/2000 with letter of 03/11/2000

Drawings, sheets:

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

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- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

- ☐ the entire international application.
- ☒ claims Nos. 40-42.

because:

- ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):
- ☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 40-42 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet
- ☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.
- ☐ no international search report has been established for the said claims Nos. .

2. A meaningful international preliminary examination report cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

- ☐ the written form has not been furnished or does not comply with the standard.
- ☐ the computer readable form has not been furnished or does not comply with the standard.

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees the applicant has:

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- ☐ restricted the claims.
- ☒ paid additional fees.
- ☐ paid additional fees under protest.
- ☐ neither restricted nor paid additional fees.
2. ☐ This Authority found that the requirement of unity of invention is not complied and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
- ☐ complied with.
- ☒ not complied with for the following reasons:
see separate sheet
4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:
- ☒ all parts.
- ☐ the parts relating to claims Nos. .

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

| | | | |
|-------------------------------|------|--------|------------------------|
| Novelty (N) | Yes: | Claims | 1a), 1c), 2-35, 38, 39 |
| | No: | Claims | 1b), 36-37, 43-46 |
| Inventive step (IS) | Yes: | Claims | |
| | No: | Claims | 1-39, 43-46 |
| Industrial applicability (IA) | Yes: | Claims | 1-39, 43-46 |
| | No: | Claims | |

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
s separate sheet

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

Although claims 40 - 42 have been drafted as separate independent claims, they relate effectively to the same subject-matter and to differ from each other only with regard to the terminology used. The application is also rendered unclear as to what concerns the invention: does the invention lie in the preparation steps or in the product itself? It is obvious from the examples in the description that the different techniques lead to different properties in the product. If the invention lies in the preparation process then a further problem of unity might arise since there is no novel inventive concept present. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it impossible to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

Hence, claims 40 - 42 do not meet the requirements of Article 6 PCT.

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

A) Invention 1:

A process for desulphurizing hydrocarbons (process claim 1 a)) with a catalyst (product claim 36) comprising a metal of group VIII, a metal of group VI, a metal oxide as carrier and a zeolite belonging to the FER type in a quantity ranging from 5 to 30 % wt.

Claim 1 a) - novelty

Claim 1 a) is a process claim for desulphurizing hydrocarbons.

The document D2 (EP-A-0 449 144) discloses a process for desulfurizing hydrocarbon oils, including Arabian Heavy fuel oil (page 10, line 26-27, about 70% of this oil boils beneath 250°C), containing sulfur (page 10, line 31: 0.15 wt%), using a catalyst

comprising metals belonging to group VIB and group VIII (page 6, line 12-30) and 2 - 35 wt% of zeolite (e.g. ZSM-35) (page 3, line 32 and page 4, line 4) into an alumina or alumina containing carrier (page 3, line 24).

The subject-matter of claim 1 therefore differs from this known in D2 in that the application relates to a process for treating hydrocarbons which boil within the range of 35 to 250 °C and the specifically chosen zeolite component ZSM-35.

Thus, the subject-matter of claim 1 a) is novel over D2 (Article 33 (2) PCT).

Claim 1 a) - inventive step

The problem to be solved by the present application is the provision of a process using a catalytic system which diminishes the sulfur content in the hydrocarbon feed and at the same time reduces to the minimum the deterioration in the octane qualities (page 2, line 10 - 15).

The solution provided by the application is the use of a catalyst comprising a zeolite belonging to the FER type in a quantity ranging from 5 to 30 %.

This catalyst system solves the problem by reducing the sulfur content and by the skeleton isomerization of the olefins present and/or by the inhibition of hydrogenation of olefinic double bonds.

Since D2 specifically names a zeolite belonging to the FER type, namely ZSM-35 (page 4, line 4), able to be used in the same process, a person skilled in the art would seriously consider using this zeolite, and since the catalyst contains the same components, the same type of reactions will take place.

Since a substantial part of the Arabian Heavy fuel oil boils beneath 250 °C, it is obvious for a person skilled in the art to use the same process for hydrocarbons which boil within the range of 35 to 250 °C.

As a result no inventive step for the subject-matter of claim 1 a) can be recognised (Article 33 (3) PCT).

Claim 36 - novelty

Claim 36 is a product-by-process claim for a catalyst.

Claims for products (a catalyst) defined in terms of a process of manufacture are

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admissible only if the products as such are new and inventive too.

The document D1 (US-A-4 808 560) discloses a catalyst comprising nickel or cobalt and molybdenum on a base selected from alumina, silica,... and having admixed 15 to less than 65 percent by weight of a crystalline zeolite (e.g. ZSM-35) (col 12, claim 1).

The document D2 discloses a catalyst comprising metals belonging to group VIB and group VIII (page 6, line 12-30) and 2 - 35 wt% of zeolite (e.g. ZSM-35) (page 3, line 32 and page 4, line 4) into an alumina or alumina containing carrier (page 3, line 24).

The subject-matter of claim 36 is further defined by means of process features, but a product is not rendered novel merely by the fact that it is produced by means of a novel process.

Thus, the catalyst as claimed in claim 36 is therefore not novel (Article 33 (2) PCT).

Claim 36 - inventive step

The problem of an inventive step will be addressed in more detail when the subject-matter of claim 36 has been rendered novel. However, since D1 and D2 disclose the same catalyst as in claim 36, the same problem will have been solved and thus no inventive step can be recognised (Article 33 (3) PCT).

Dependent claims 3 - 28, 31, 37, 42 - 45

A positive preliminary examination report for the dependent claims 3-28, 31, 42-45 can only be established when the requirements concerning novelty and inventive step for the independent claims are met .

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B) Invention 2:

A process for desulphurizing hydrocarbons (process claim 1 b)) and catalysts (product claims 38 to 39) comprising a metal of group VIII, a metal of group VI, a metal oxide as carrier and phosphorous in a quantity ranging from 0.1 to 10 % wt.

Claim 1 b) - novelty

Claim 1 b) is a process claim for desulfurizing hydrocarbons.

Document D3 (US-A-5576256) (cited in the search report and in the application) discloses a process for desulphurizing and improving the octane number of hydrocarbons (col 2, line 11-22) having a sulfur content exceeding 1000 ppm (col 6, line 9-10), with skeleton isomerization (col 2, line 23), using a catalyst (col 2, line 49), which comprises a metal of group VIII (col 3, line 4-5), a metal of group VI (col 3, line 5-6), an alumina matrix (col 2, line 64), phosphorous (col 3, line 7) in a quantity ranging from 0.1 - 10 % weight catalyst (col 7, table 1), said catalyst is obtained by impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 (col 4, line 53-55), with an aqueous solution of the metal of group VIII (col 4, line 56) and group VI (col 4, line 55).

Document D4 discloses a process for desulfurizing gas oils with a boiling point range from 150 to 400 °C (page 6, line 47, specifically named endpoints of overlapping ranges are novelty destroying) having a sulfur content of not more than 2 wt% of sulfur (page 6, line 49) using a catalyst in which an inorganic oxide carrier (page 3, line 57), is impregnated with a group VIII metal, a group VI metal and phosphorous (page 3, line 58) in an amount of 0.1 to 15 wt% in terms of oxide (page 4, line 2).

Thus the subject-matter of claim 1 b) has already been anticipated by the prior art D3 and D4 and is therefore not novel (Article 33 (2) PCT).

Claim 1 b) - inventive step

The problem to be solved may be regarded as "How to provide an alternative process for desulfurizing hydrocarbons with very low losses of RON and MON".

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The solution proposed in claim 1 b) of the present application cannot be considered as involving an inventive step (Article 33 (3) PCT) for the following reasons. D3 is concerned with the same technical problems, notably to desulfurize a hydrocarbon feed with very low losses of RON and MON, and since D3 solves the problem in the same manner, no inventive step can be recognised.

Claim 43 - 45 - novelty

Claims for products (a catalyst) defined in terms of a process of manufacture are admissible only if the products as such are new and inventive too.

Claims 43 to 45 are product claims for the same catalyst defined in terms of alternative processes.

Document D4 (EP-A-0665280) is considered to represent the closest prior art to the subject-matter of claim 43 to 45 and discloses a catalyst (page 4, line 7) comprising a metal of group VIII (page 4, line 11), a metal of group VI (page 4, line 10), a metal oxide as carrier (page 4, line 8) and P in a quantity ranging from 2 to 15 wt% (page 5, line 35-37).

The fact that the products may have been produced by a possibly novel process cannot be considered as limiting.

Thus the subject-matter of claims 43 to 45 has already been anticipated by the prior art D4 and is therefore not novel (Article 33 (2) PCT).

Claim 43 - 45 - inventive step

The problem to be solved may be regarded as "How to provide an alternative catalyst suitable for desulfurizing hydrocarbons".

The solution proposed in claims 43 to 45 of the present application cannot be considered as involving an inventive step (Article 33 (3) PCT) for the following reasons. D4 is concerned with the same technical problems, notably to synthesize a catalyst for desulfurizing a hydrocarbon feed, and since D4 solves the problem in the same manner, no inventive step can be recognised.

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Claim 46 - novelty and inventive step

D4 further discloses in specific embodiments amounts of phosphorous between 1 and 5 wt% (page 15).

Thus, the subject-matter of claim 46 lacks novelty (Article 33 (2) PCT).

With regard to inventive step, the reasons set out above for claim 43 to 45 also apply for claim 46. Thus, no inventive step can be recognised (Article 33 (3) PCT).

Dependent claims 3 - 13, 29 - 31, 42 - 45

A positive preliminary examination report for the dependent claims 3-28, 31, 42-45 can only be established when the requirements concerning novelty and inventive step for the independent claims are complying.

C) Claim 1 c) and claim 38:

Claim 1 c) is a further permutation of claim 1 a) or claim 1 b). For the purpose of this examination 1 c) can be regarded as a dependent claim. This also applies to claim 38.

Claim 1 c) - novelty and inventive step

Claim 1 c) is a process for desulfurizing hydrocarbons with a catalyst containing a zeolite and phosphorous.

D4 discloses a process for preparing a catalyst for hydrodesulfurization in which an inorganic oxide carrier containing a crystalline aluminosilicate can be impregnated with a Group Vi metal, a Group VIII metal and phosphoric acid, to obtain a phosphorous content of 0.1 - 15 wt% in terms of oxide. The crystalline aluminosilicate includes ZSM type zeolite in an amount of 2 to 15 wt% (page 3, line 56-58 and page 4 line 1-3 and 27-37).

Document D4, which is considered to represent the most relevant state of the art, discloses ZSM type zeolites from which the subject-matter of claim 1 c) differs in that a specific type of zeolite (FER) is comprised.

The subject-matter of claim 1 c) is therefore novel (Article 33(2) PCT).

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For inventive step, the documents D1 (col 5, line 32), D2 (page 4, line 4), D5 (US-A-5378352)(col 6, line 37) and D6 (US-A-4629717)(col 8, line 2) must be regarded. These all disclose a process for desulfurizing a hydrocarbon feed with a catalyst including a FER type zeolite. For a person skilled in the art it thus would be obvious to use a zeolite of the FER type (ZSM-35) for a process as claimed in 1 c). Thus, no inventive step can be recognised for the subject-matter of claim 1 c) (Article 33 (3) PCT).

Claim 38 - novelty

Claim 38 is drafted as an independent product claim for a catalyst.

Document D4 (EP-A-0665280) is considered to represent the closest prior art to the subject-matter of claim 33 and discloses a catalyst (page 4, line 7) comprising a metal of group VIII (page 4, line 11), a metal of group VI (page 4, line 10), a metal oxide as carrier (page 4, line 8) and a zeolite of the ZSM group (page 4, line 8), in a quantity ranging from 2 to 15 wt% (page 4, line 37) and phosphorous in a quantity ranging from 0.1 to 10 % (page 4, line 11).

The catalyst as claimed in claim 38 has the additional feature that the zeolite needs to belong to the FER type and is therefore novel (Article 33 (2) PCT).

Claim 38 inventive step:

The subject-matter of claim 38 contains the additional technical feature in view of D4, that the zeolite needs to belong to the FER type, this includes ZSM-35 according to the description. ZSM-35 is merely one of several straightforward possibilities from which the skilled person would select, in accordance with the contents of D4 (page 4, line 29), without the exercise of inventive skill, in order to solve the problem posed. Furthermore the use of ZSM-35 or FER type zeolites for use in a catalyst to desulphurize hydrocarbons has already been stated in D1 (page 4, line 35-45), D2 (page 4, line 4), D5 (US-A-5378352) (cited in the search report and in the application) (page 6, line 37), and D6 (US-A-4629717) (page 8, line 2 and 5).

In view of the paragraph above, the skilled person would regard it obvious to combine all the features set out in claim 38.

Thus, the subject-matter of claim 38 does not involve an inventive step and does not

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satisfy the criterion set forth in Article 33 (3) PCT.

Claim 39 - novelty and inventive step

Since claim 39 is dependent on claim 38, which is novel, the subject-matter of claim 39 can be considered novel too (Article 33 (2) PCT).

With regard to inventive step, the reasons set out above for claim 38 also apply for claim 39. Thus, no inventive step can be recognised (Article 33 (3) PCT).

Re Item VII

Certain defects in the international application

Claim 38 is drafted as an independent claim but effectively relates back to the catalyst mentioned in claim 1 c). Therefore, it should be drafted as a dependent claim.

If the claim relates to a product as such which is new and inventive, a problem of unity arises.

Claims 43 - 45 have been drafted as separate independent claims but they relate effectively to the same subject-matter (the catalyst from invention 2) and to differ from each other only with regard to the terminology used. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it impossible to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

Hence, claims 43 - 45 do not meet the requirements of Article 6 PCT.



30) The process according to claim 1 wherein the catalyst contains phosphorous as component A and is prepared by impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 followed by
5 impregnation with an aqueous solution of the metal of group VIII and an aqueous solution of the metal of group VI, wherein the metal oxide carrier has a surface area lower than $240 \text{ m}^2/\text{g}$.

31) The process according to claim 1 or 2 carried out
10 at a temperature ranging from 220 to 340°C , at a pressure ranging from 5 to 20 Kg/cm^2 , at a LHSV ranging from 1 to 10 h^{-1} in the presence of hydrogen in a quantity ranging from 100 to 500 times with respect to the hydrocarbons present (Nl/l).

32) A catalyst containing a metal of group VIII, a
15 metal of group VI, a metal oxide as carrier and a zeolite belonging to the FER type, in a quantity ranging from 5 to 30% by weight with respect to the total weight of the catalyst, prepared as follows:

20 a) an alcohol dispersion is prepared, containing a soluble salt of the metal of group VIII, the zeolite of the FER group and an organic source of Aluminum;

b) an aqueous solution is prepared, containing a soluble salt of the metal of group VI and optionally formamide;

c) the alcohol dispersion and the aqueous solution are mixed, obtaining a gel;

d) aging of the gel at a temperature ranging from 10 to 40°C;

e) drying of the gel;

f) calcination of the gel.

33) A catalyst which comprise a metal of group VIII, a metal of group VI, a metal oxide as carrier, a zeolite of the FER type, in a quantity ranging from 5 to 30% by weight with respect to the total weight of the catalyst, and phosphorous in a quantity ranging from 0.1 to 10%.

34) A process for preparing the catalyst of claim 33 comprising the following steps:

a) an alcohol dispersion is prepared, containing a soluble salt of the metal of group VIII, the zeolite of FER type and an organic source of aluminum;



- b) an aqueous solution is prepared, containing a soluble salt of the metal of group VI, H_3PO_4 and optionally formamide;
- c) the alcohol dispersion and the aqueous solution are mixed, obtaining a gel;
- d) aging of the gel at a temperature ranging from 10 to 40°C;
- e) drying of the gel;
- f) calcination of the gel.
- 35) A process for preparing the catalyst of claim 33 comprising the following steps:
- a) an alcohol dispersion is prepared, containing a soluble salt of the metal of group VIII, a zeolite of FER type and an organic source of aluminum;
- b) an aqueous solution is prepared, containing a soluble salt of the metal of group VI, and optionally formamide;
- c) the alcohol dispersion and the aqueous solution are mixed, obtaining a gel;
- d) aging of the gel at a temperature ranging from 10 to 40°C;
- e) drying of the gel;

f) calcination of the gel.

g) impregnation of the catalyst obtained from step f) with an aqueous solution of H_3PO_4 , drying and calcination.

5 36) Process for preparing the catalyst of claim 33 comprising:

a) impregnation of metal oxide with an aqueous solution of H_3PO_4 , drying and calcination,

b) impregnation of the material resulting from step a) with an aqueous solution of metal of group VIII and an aqueous solution of metal of group VI,

c) drying and calcination of the material resulting from step b)

d) mixing the impregnated metal oxide obtained from step c) with the zeolite of FER type.

37) The catalyst according to claim 33 containing phosphorous in a quantity ranging from 1 to 5 % wt with respect to the total weight of the catalyst.

38) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by

impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 , followed by impregnation with an aqueous solution of the metal of group VIII and an aqueous solution of the metal of group VI.

5 39) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by drying and calcination of a gel obtained mixing an alcohol
10 dispersion containing a soluble salt of the metal of group VIII and an organic source of aluminum with an aqueous solution containing a soluble salt of the metal of group VI and H_3PO_4 .

15 40) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by impregnation with an aqueous solution of H_3PO_4 of a gel, dried and calcined, obtained mixing an alcohol
20 dispersion containing a soluble salt of the metal of group VIII and an organic source of aluminum with an

aqueous solution containing a soluble salt of the metal of group VI.

5 41) The catalyst according to claim 38, 39 or 40 containing phosphorous in a quantity ranging from 1 to 5 % wt with respect to the total weight of the catalyst.

42) The process according to claim 1 or 2 wherein the hydrocarbons boiling within the range of 35 to 250°C contain more than 1000 ppm of S.

10 43) The process according to claim 1 or 2 wherein hydrocarbons boiling within the range of 35 to 250°C derive from cracking processes.

44) The process according to claim 1 or 2 wherein the catalyst is in extruded form.

15 45) The process according to claim 31 carried out at a temperature ranging from 220 to 330°C.



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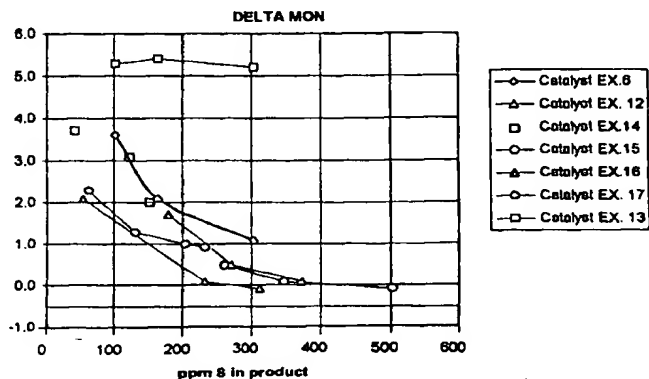
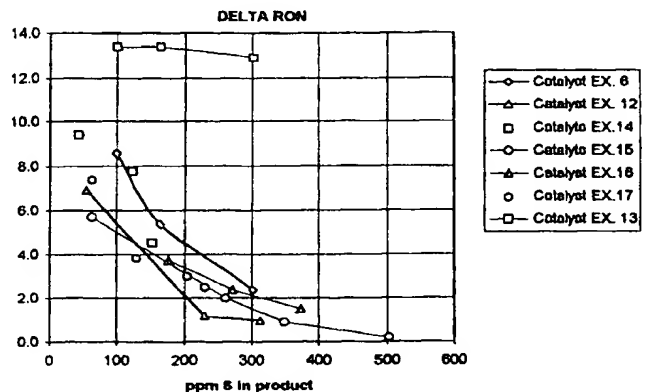
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| (21) International Application Number: PCT/EP99/05577 (22) International Filing Date: 3 August 1999 (03.08.99) (30) Priority Data: 98202744.3 15 August 1998 (15.08.98) EP (71) Applicants (for all designated States except US): ENITEC-NOLOGIE S.P.A. [IT/IT]; Via F. Maritano, 26, I-20097 San Donato Milanese (IT). REPSOL PETROLEO S.A. [ES/ES]; C/Embajadores, 183, E-28045 Madrid (ES). ELF ANTAR FRANCE S.A. [FR/FR]; Tour Elf, 2, place de la Coupole, La Defense 6, F-92400 Courbevoie (FR). EURON S.P.A. [IT/IT]; Via F. Maritano, 26, I-20097 San Donato Milanese (IT). AGIP PETROLI S.P.A. [IT/IT]; Via Laurentina, 449, I-00198 Rome (IT). (72) Inventors; and (75) Inventors/Applicants (for US only): ZANIBELLI, Laura [IT/IT]; Via. F. Donati, 1, I-26013 Crema (IT). AR-RIGONI, Virginio [IT/IT]; Via A. Cossa, 2, I-20138 Milan (IT). ALBERTOS, Fernando [ES/ES]; C/Palma 50, 3° Derecha, E-28015 Madrid (ES). ATANES, Evangelina [ES/ES]; Plaza de Penuelas 5, 3° C, E-28005 Madrid (ES). CHOLLEY, Thierry [FR/FR]; 230, rue Vendome, F-69003 | | Lyon (FR). PANARELLO, Febronio [IT/IT]; Via XXV Aprile, 11/D, I-20061 Carugate (IT). (74) Agents: DE GREGORI, Antonella et al.; Ing. Barzanò & Zanardo, Milano S.p.A., Via Borgonuovo, 10, I-20121 Milan (IT). (81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). |

Published

With international search report.

(54) Title: PROCESS AND CATALYSTS FOR UPGRADING OF HYDROCARBONS BOILING IN THE NAPHTHA RANGE**(57) Abstract**

The present invention relates to the use of a catalytic system comprising a metal of group VIII, a metal of group VI, a metal oxide as carrier and suitable quantities of a component selected from a zeolite of the FER type, phosphorous, and a mixture thereof, in upgrading of hydrocarbons boiling in the naphtha range containing sulfur impurities, namely in hydrodesulfurization with contemporaneous skeleton isomerization of olefins contained in said hydrocarbons and/or with reduction of olefins hydrogenation, carried out in a single step.



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| BG | Bulgaria | HU | Hungary | ML | Mali | TT | Trinidad and Tobago |
| BJ | Benin | IE | Ireland | MN | Mongolia | UA | Ukraine |
| BR | Brazil | IL | Israel | MR | Mauritania | UG | Uganda |
| BY | Belarus | IS | Iceland | MW | Malawi | US | United States of America |
| CA | Canada | IT | Italy | MX | Mexico | UZ | Uzbekistan |
| CF | Central African Republic | JP | Japan | NE | Niger | VN | Viet Nam |
| CG | Congo | KE | Kenya | NL | Netherlands | YU | Yugoslavia |
| CH | Switzerland | KG | Kyrgyzstan | NO | Norway | ZW | Zimbabwe |
| CI | Côte d'Ivoire | KP | Democratic People's Republic of Korea | NZ | New Zealand | | |
| CM | Cameroon | | | PL | Poland | | |
| CN | China | KR | Republic of Korea | PT | Portugal | | |
| CU | Cuba | KZ | Kazakstan | RO | Romania | | |
| CZ | Czech Republic | LC | Saint Lucia | RU | Russian Federation | | |
| DE | Germany | LI | Liechtenstein | SD | Sudan | | |
| DK | Denmark | LK | Sri Lanka | SE | Sweden | | |
| EE | Estonia | LR | Liberia | SG | Singapore | | |

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/EP 99/05577

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C10G45/12 C10G45/64 C10G45/08 B01J23/85 B01J29/69
B01J27/185 B01J27/188

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C10G B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|---|---|
| X | US 5 378 352 A (DEGNAN THOMAS F ET AL) 3 January 1995 (1995-01-03) cited in the application | 1,3-15, 42 |
| A | claims 1-3,6-10,14-16 column 5, line 1 - line 14 column 5, line 43 -column 6, line 10 column 6, line 32 - line 34 table 1 | 32,43 |
| X | US 5 576 256 A (GARCIA WOLFGANG ET AL) 19 November 1996 (1996-11-19) cited in the application | 1-8,10, 12,13, 18,29, 31,38-45 |
| | column 2, line 64 -column 3, line 23 column 5, line 57 -column 6, line 43 examples 2,4-7 | |
| | --- -/-- | |

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

Z document member of the same patent family

Date of the actual completion of the international search

10 November 1999

Date of mailing of the international search report

24.11.99

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
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Fax: (+31-70) 340-3016

Authorized officer

Zuurdeeg, B

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category ° | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|--|
| X A | US 4 629 717 A (CHAO TAI-HSIANG) 16 December 1986 (1986-12-16) claims 1,4,5,10,11,15 column 7, line 64 -column 8, line 35 column 9, line 8 - line 21 example VI --- | 1,38-41 1,5-13, 29-31, 33,36,37 |
| X | EP 0 448 117 A (COSMO SOGO KENKYUSHO KK ;COSMO OIL CO LTD (JP)) 25 September 1991 (1991-09-25) claims 1-3,6,7,9,18 page 6, line 29 - line 35 page 7, line 54 -page 8, line 1 page 8, line 17 - line 21 example 8 --- | 1,38-41 |
| A | US 5 482 617 A (COLLINS NICK A ET AL) 9 January 1996 (1996-01-09) claims 1-6,8-10,13-16,18-25 column 5, line 21 - line 42 --- | 1,3-5, 12-16, 25,31, 43,45 |
| A | EP 0 449 144 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 October 1991 (1991-10-02) claims 1,3,8,10,11,17-19-22,24 page 5, line 20 -page 6, line 38 page 9, line 34 - line 44 tables 1-3,5 --- | 1,3-15, 31,32 |
| A | EP 0 665 280 A (COSMO OIL CO LTD ;PETROLEUM ENERGY CENTER FOUND (JP)) 2 August 1995 (1995-08-02) the whole document --- | 1-8,12, 13,33,37 |
| A | US 4 808 560 A (OLECK STEPHEN M) 28 February 1989 (1989-02-28) claims 1,2,4,7,8 column 6, line 23 - line 68 --- | 1,3-16 |
| A | EP 0 442 159 A (SHELL INT RESEARCH) 21 August 1991 (1991-08-21) cited in the application claims 1-9 page 5, line 55 -page 6, line 17 --- | 1,25,32 |
| | -/-- | |

INTERNATIONAL SEARCH REPORT

Patent Application No

PCT/EP 99/05577

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|------------|--|-----------------------|
| A | <p> DATABASE WPI Section Ch, Week 8630 Derwent Publications Ltd., London, GB; Class H04, AN 86-193476 XP002089923 & JP 61 126196 A (JUSHITSUYU TAISAKU GIJUTSU), 13 June 1986 (1986-06-13) abstract ----- </p> | |

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP 99/05577

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/EP 99/05577

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1a,c-29,31-37,42-45

Catalyst containing FER type zeolite

2. Claims: 1b,30,38-41

Catalyst containing phosphorous

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 99/05577

| Patent document cited in search report | | Publication date | Patent family member(s) | Publication date |
|---|---|---------------------|---|--|
| US 5378352 | A | 03-01-1995 | AU 658937 B AU 2734092 A CA 2081758 A DE 69222396 D DE 69222396 T EP 0543529 A ES 2106833 T JP 5247474 A | 04-05-1995 20-05-1993 20-05-1993 30-10-1997 26-02-1998 26-05-1993 16-11-1997 24-09-1993 |
| US 5576256 | A | 19-11-1996 | BR 9502505 A DE 19518607 A FR 2720073 A GB 2289689 A,B GB 2323094 A,B NL 1000428 C NL 1000428 A US 5591324 A US 5770047 A | 27-02-1996 30-11-1995 24-11-1995 29-11-1995 16-09-1998 24-12-1996 23-11-1995 07-01-1997 23-06-1998 |
| US 4629717 | A | 16-12-1986 | US 4727209 A | 23-02-1988 |
| EP 0448117 | A | 25-09-1991 | JP 2789489 B JP 3275142 A JP 4007044 A DE 69104247 D DE 69104247 T US 5182250 A US 5244858 A | 20-08-1998 05-12-1991 10-01-1992 03-11-1994 09-03-1995 26-01-1993 14-09-1993 |
| US 5482617 | A | 09-01-1996 | NONE | |
| EP 0449144 | A | 02-10-1991 | JP 2547115 B JP 3284354 A DE 69103058 D US 5187133 A | 23-10-1996 16-12-1991 01-09-1994 16-02-1993 |
| EP 0665280 | A | 02-08-1995 | JP 7197038 A JP 2875148 B JP 7194984 A | 01-08-1995 24-03-1999 01-08-1995 |
| US 4808560 | A | 28-02-1989 | NONE | |
| EP 0442159 | A | 21-08-1991 | US 5043520 A DE 69009727 D DE 69009727 T | 27-08-1991 14-07-1994 22-09-1994 |
| JP 61126196 | A | 13-06-1986 | JP 1902513 C JP 6031333 B | 08-02-1995 27-04-1994 |

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

| | | |
|---|---|--|
| Applicant's or agent's file reference Cal 75859 | FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416) | |
| International application No. PCT/EP99/05577 | International filing date (day/month/year) 03/08/1999 | Priority date (day/month/year) 15/08/1998 |
| International Patent Classification (IPC) or national classification and IPC C10G45/12 | | |
| Applicant ENITECNOLOGIE, S.P.A. et al. | | |

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 12 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☒ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

| | |
|---|---|
| Date of submission of the demand 04/01/2000 | Date of completion of this report 04.12.2000 |
| Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465 | Authorized officer de Cauwer, R Telephone No. +49 89 2399 7344  |

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/05577

I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:

Description, pages:

1-52 as originally filed

Claims, No.:

1-29 as originally filed

30-46 as received on 06/11/2000 with letter of 03/11/2000

Drawings, sheets:

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/EP99/05577

- ☐ the description, pages:
☐ the claims, Nos.:
☐ the drawings, sheets:
5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):
(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

- ☐ the entire international application.
☒ claims Nos. 40-42.

because:

- ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):
- ☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 40-42 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet
- ☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.
- ☐ no international search report has been established for the said claims Nos. .
2. A meaningful international preliminary examination report cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:
- ☐ the written form has not been furnished or does not comply with the standard.
☐ the computer readable form has not been furnished or does not comply with the standard.

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees the applicant has:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP99/05577

- ☐ restricted the claims.
 - ☒ paid additional fees.
 - ☐ paid additional fees under protest.
 - ☐ neither restricted nor paid additional fees.
2. ☐ This Authority found that the requirement of unity of invention is not complied and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
- ☐ complied with.
 - ☒ not complied with for the following reasons:
see separate sheet
4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:
- ☒ all parts.
 - ☐ the parts relating to claims Nos. .

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

| | | |
|-------------------------------|------|-------------------------------|
| Novelty (N) | Yes: | Claims 1a), 1c), 2-35, 38, 39 |
| | No: | Claims 1b), 36-37, 43-46 |
| Inventive step (IS) | Yes: | Claims |
| | No: | Claims 1-39, 43-46 |
| Industrial applicability (IA) | Yes: | Claims 1-39, 43-46 |
| | No: | Claims |

2. Citations and explanations **see separate sheet**

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

Although claims 40 - 42 have been drafted as separate independent claims, they relate effectively to the same subject-matter and to differ from each other only with regard to the terminology used. The application is also rendered unclear as to what concerns the invention: does the invention lie in the preparation steps or in the product itself? It is obvious from the examples in the description that the different techniques lead to different properties in the product. If the invention lies in the preparation process then a further problem of unity might arise since there is no novel inventive concept present. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it impossible to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

Hence, claims 40 - 42 do not meet the requirements of Article 6 PCT.

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

A) Invention 1:

A process for desulphurizing hydrocarbons (process claim 1 a)) with a catalyst (product claim 36) comprising a metal of group VIII, a metal of group VI, a metal oxide as carrier and a zeolite belonging to the FER type in a quantity ranging from 5 to 30 % wt.

Claim 1 a) - novelty

Claim 1 a) is a process claim for desulphurizing hydrocarbons.

The document D2 (EP-A-0 449 144) discloses a process for desulfurizing hydrocarbon oils, including Arabian Heavy fuel oil (page 10, line 26-27, about 70% of this oil boils beneath 250°C) , containing sulfur (page 10, line 31: 0.15 wt%), using a catalyst

comprising metals belonging to group VIB and group VIII (page 6, line 12-30) and 2 - 35 wt% of zeolite (e.g. ZSM-35) (page 3, line 32 and page 4, line 4) into an alumina or alumina containing carrier (page 3, line 24).

The subject-matter of claim 1 therefore differs from this known in D2 in that the application relates to a process for treating hydrocarbons which boil within the range of 35 to 250 °C and the specifically chosen zeolite component ZSM-35.

Thus, the subject-matter of claim 1 a) is novel over D2 (Article 33 (2) PCT).

Claim 1 a) - inventive step

The problem to be solved by the present application is the provision of a process using a catalytic system which diminishes the sulfur content in the hydrocarbon feed and at the same time reduces to the minimum the deterioration in the octane qualities (page 2, line 10 - 15).

The solution provided by the application is the use of a catalyst comprising a zeolite belonging to the FER type in a quantity ranging from 5 to 30 %.

This catalyst system solves the problem by reducing the sulfur content and by the skeleton isomerization of the olefins present and/or by the inhibition of hydrogenation of olefinic double bonds.

Since D2 specifically names a zeolite belonging to the FER type, namely ZSM-35 (page 4, line 4), able to be used in the same process, a person skilled in the art would seriously consider using this zeolite, and since the catalyst contains the same components, the same type of reactions will take place.

Since a substantial part of the Arabian Heavy fuel oil boils beneath 250 °C, it is obvious for a person skilled in the art to use the same process for hydrocarbons which boil within the range of 35 to 250 °C.

As a result no inventive step for the subject-matter of claim 1 a) can be recognised (Article 33 (3) PCT).

Claim 36 - novelty

Claim 36 is a product-by-process claim for a catalyst.

Claims for products (a catalyst) defined in terms of a process of manufacture are

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International application No. PCT/EP99/05577

admissible only if the products as such are new and inventive too.

The document D1 (US-A-4 808 560) discloses a catalyst comprising nickel or cobalt and molybdenum on a base selected from alumina, silica,... and having admixed 15 to less than 65 percent by weight of a crystalline zeolite (e.g. ZSM-35) (col 12, claim 1).

The document D2 discloses a catalyst comprising metals belonging to group VIB and group VIII (page 6, line 12-30) and 2 - 35 wt% of zeolite (e.g. ZSM-35) (page 3, line 32 and page 4, line 4) into an alumina or alumina containing carrier (page 3, line 24).

The subject-matter of claim 36 is further defined by means of process features, but a product is not rendered novel merely by the fact that it is produced by means of a novel process.

Thus, the catalyst as claimed in claim 36 is therefore not novel (Article 33 (2) PCT).

Claim 36 - inventive step

The problem of an inventive step will be addressed in more detail when the subject-matter of claim 36 has been rendered novel. However, since D1 and D2 disclose the same catalyst as in claim 36, the same problem will have been solved and thus no inventive step can be recognised (Article 33 (3) PCT).

Dependent claims 3 - 28, 31, 37, 42 - 45

A positive preliminary examination report for the dependent claims 3-28, 31, 42-45 can only be established when the requirements concerning novelty and inventive step for the independent claims are met .

B) Invention 2:

A process for desulphurizing hydrocarbons (process claim 1 b)) and catalysts (product claims 38 to 39) comprising a metal of group VIII, a metal of group VI, a metal oxide as carrier and phosphorous in a quantity ranging from 0.1 to 10 % wt.

Claim 1 b) - novelty

Claim 1 b) is a process claim for desulfurizing hydrocarbons.

Document D3 (US-A-5576256) (cited in the search report and in the application) discloses a process for desulphurizing and improving the octane number of hydrocarbons (col 2, line 11-22) having a sulfur content exceeding 1000 ppm (col 6, line 9-10), with skeleton isomerization (col 2, line 23), using a catalyst (col 2, line 49), which comprises a metal of group VIII (col 3, line 4-5), a metal of group VI (col 3, line 5-6), an alumina matrix (col 2, line 64), phosphorous (col 3, line 7) in a quantity ranging from 0.1 - 10 % weight catalyst (col 7, table 1), said catalyst is obtained by impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 (col 4, line 53-55), with an aqueous solution of the metal of group VIII (col 4, line 56) and group VI (col 4, line 55).

Document D4 discloses a process for desulfurizing gas oils with a boiling point range from 150 to 400 °C (page 6, line 47, specifically named endpoints of overlapping ranges are novelty destroying) having a sulfur content of not more than 2 wt% of sulfur (page 6, line 49) using a catalyst in which an inorganic oxide carrier (page 3, line 57), is impregnated with a group VIII metal, a group VI metal and phosphorous (page 3, line 58) in an amount of 0.1 to 15 wt% in terms of oxide (page 4, line 2).

Thus the subject-matter of claim 1 b) has already been anticipated by the prior art D3 and D4 and is therefore not novel (Article 33 (2) PCT).

Claim 1 b) - inventive step

The problem to be solved may be regarded as "How to provide an alternative process for desulfurizing hydrocarbons with very low losses of RON and MON".

The solution proposed in claim 1 b) of the present application cannot be considered as involving an inventive step (Article 33 (3) PCT) for the following reasons. D3 is concerned with the same technical problems, notably to desulfurize a hydrocarbon feed with very low losses of RON and MON, and since D3 solves the problem in the same manner, no inventive step can be recognised.

Claim 43 - 45 - novelty

Claims for products (a catalyst) defined in terms of a process of manufacture are admissible only if the products as such are new and inventive too.

Claims 43 to 45 are product claims for the same catalyst defined in terms of alternative processes.

Document D4 (EP-A-0665280) is considered to represent the closest prior art to the subject-matter of claim 43 to 45 and discloses a catalyst (page 4, line 7) comprising a metal of group VIII (page 4, line 11), a metal of group VI (page 4, line 10), a metal oxide as carrier (page 4, line 8) and P in a quantity ranging from 2 to 15 wt% (page 5, line 35-37).

The fact that the products may have been produced by a possibly novel process cannot be considered as limiting.

Thus the subject-matter of claims 43 to 45 has already been anticipated by the prior art D4 and is therefore not novel (Article 33 (2) PCT).

Claim 43 - 45 - inventive step

The problem to be solved may be regarded as "How to provide an alternative catalyst suitable for desulfurizing hydrocarbons".

The solution proposed in claims 43 to 45 of the present application cannot be considered as involving an inventive step (Article 33 (3) PCT) for the following reasons. D4 is concerned with the same technical problems, notably to synthesize a catalyst for desulfurizing a hydrocarbon feed, and since D4 solves the problem in the same manner, no inventive step can be recognised.

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Claim 46 - novelty and inventive step

D4 further discloses in specific embodiments amounts of phosphorous between 1 and 5 wt% (page 15).

Thus, the subject-matter of claim 46 lacks novelty (Article 33 (2) PCT).

With regard to inventive step, the reasons set out above for claim 43 to 45 also apply for claim 46. Thus, no inventive step can be recognised (Article 33 (3) PCT).

Dependent claims 3 - 13, 29 - 31, 42 - 45

A positive preliminary examination report for the dependent claims 3-28, 31, 42-45 can only be established when the requirements concerning novelty and inventive step for the independent claims are complying.

C) Claim 1 c) and claim 38:

Claim 1 c) is a further permutation of claim 1 a) or claim 1 b). For the purpose of this examination 1 c) can be regarded as a dependent claim. This also applies to claim 38.

Claim 1 c) - novelty and inventive step

Claim 1 c) is a process for desulfurizing hydrocarbons with a catalyst containing a zeolite and phosphorous.

D4 discloses a process for preparing a catalyst for hydrodesulfurization in which an inorganic oxide carrier containing a crystalline aluminosilicate can be impregnated with a Group Vi metal, a Group VIII metal and phosphoric acid, to obtain a phosphorous content of 0.1 - 15 wt% in terms of oxide. The crystalline aluminosilicate includes ZSM type zeolite in an amount of 2 to 15 wt% (page 3, line 56-58 and page 4 line 1-3 and 27-37).

Document D4, which is considered to represent the most relevant state of the art, discloses ZSM type zeolites from which the subject-matter of claim 1 c) differs in that a specific type of zeolite (FER) is comprised.

The subject-matter of claim 1 c) is therefore novel (Article 33(2) PCT).

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International application No. PCT/EP99/05577

For inventive step, the documents D1 (col 5, line 32), D2 (page 4, line 4), D5 (US-A-5378352)(col 6, line 37) and D6 (US-A-4629717)(col 8, line 2) must be regarded. These all disclose a process for desulfurizing a hydrocarbon feed with a catalyst including a FER type zeolite. For a person skilled in the art it thus would be obvious to use a zeolite of the FER type (ZSM-35) for a process as claimed in 1 c). Thus, no inventive step can be recognised for the subject-matter of claim 1 c) (Article 33 (3) PCT).

Claim 38 - novelty

Claim 38 is drafted as an independent product claim for a catalyst.

Document D4 (EP-A-0665280) is considered to represent the closest prior art to the subject-matter of claim 33 and discloses a catalyst (page 4, line 7) comprising a metal of group VIII (page 4, line 11), a metal of group VI (page 4, line 10), a metal oxide as carrier (page 4, line 8) and a zeolite of the ZSM group (page 4, line 8), in a quantity ranging from 2 to 15 wt% (page 4, line 37) and phosphorous in a quantity ranging from 0.1 to 10 % (page 4, line 11).

The catalyst as claimed in claim 38 has the additional feature that the zeolite needs to belong to the FER type and is therefore novel (Article 33 (2) PCT).

Claim 38 inventive step:

The subject-matter of claim 38 contains the additional technical feature in view of D4, that the zeolite needs to belong to the FER type, this includes ZSM-35 according to the description. ZSM-35 is merely one of several straightforward possibilities from which the skilled person would select, in accordance with the contents of D4 (page 4, line 29), without the exercise of inventive skill, in order to solve the problem posed. Furthermore the use of ZSM-35 or FER type zeolites for use in a catalyst to desulphurize hydrocarbons has already been stated in D1 (page 4, line 35-45), D2 (page 4, line 4), D5 (US-A-5378352) (cited in the search report and in the application) (page 6, line 37), and D6 (US-A-4629717) (page 8, line 2 and 5).

In view of the paragraph above, the skilled person would regard it obvious to combine all the features set out in claim 38.

Thus, the subject-matter of claim 38 does not involve an inventive step and does not

satisfy the criterion set forth in Article 33 (3) PCT.

Claim 39 - novelty and inventive step

Since claim 39 is dependent on claim 38, which is novel, the subject-matter of claim 39 can be considered novel too (Article 33 (2) PCT).

With regard to inventive step, the reasons set out above for claim 38 also apply for claim 39. Thus, no inventive step can be recognised (Article 33 (3) PCT).

Re Item VII

Certain defects in the international application

Claim 38 is drafted as an independent claim but effectively relates back to the catalyst mentioned in claim 1 c). Therefore, it should be drafted as a dependent claim.

If the claim relates to a product as such which is new and inventive, a problem of unity arises.

Claims 43 - 45 have been drafted as separate independent claims but they relate effectively to the same subject-matter (the catalyst from invention 2) and to differ from each other only with regard to the terminology used. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it impossible to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

Hence, claims 43 - 45 do not meet the requirements of Article 6 PCT.

30) The process according to claim 1 wherein the catalyst contains phosphorous as component A and is prepared by impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 followed by impregnation with an aqueous solution of the metal of group VIII and an aqueous solution of the metal of group VI, wherein the metal oxide carrier has a surface area lower than $240 \text{ m}^2/\text{g}$.

31) The process according to claim 1 or 2 carried out at a temperature ranging from 220 to 340°C , at a pressure ranging from 5 to 20 Kg/cm^2 , at a LHSV ranging from 1 to 10 h^{-1} in the presence of hydrogen in a quantity ranging from 100 to 500 times with respect to the hydrocarbons present (Nl/l).

~~32) A catalyst containing a metal of group VIII, a metal of group VI, a metal oxide as carrier and a zeolite belonging to the FER type, in a quantity ranging from 5 to 30% by weight with respect to the total weight of the catalyst, prepared as follows:~~

~~a) an alcohol dispersion is prepared, containing a soluble salt of the metal of group VIII, the zeolite of the FER group and an organic source of Aluminum;~~

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~~aqueous solution containing a soluble salt of the metal of group VI.~~

41) The catalyst according to claim 38, 39 or 40 containing phosphorous in a quantity ranging from 1 to 5 % wt with respect to the total weight of the catalyst.

32 42) The process according to claim 1 or 2 wherein the hydrocarbons boiling within the range of 35 to 250°C contain more than 1000 ppm of S.

10 33 43) The process according to claim 1 or 2 wherein hydrocarbons boiling within the range of 35 to 250°C derive from cracking processes.

34 44) The process according to claim 1 or 2 wherein the catalyst is in extruded form.

15 35 45) The process according to claim 31 carried out at a temperature ranging from 220 to 330°C.

36 32) A catalyst containing a metal of group VIII, a metal of group VI, a metal oxide as carrier and a zeolite belonging to the FER type, in a quantity ranging from 5 to 30% by weight with respect to the total weight of the catalyst, prepared as follows:

a) an alcohol dispersion is prepared, containing a soluble salt of the metal of group VIII, the zeolite of the FER group and an organic source of Aluminum;

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b) an aqueous solution is prepared, containing a soluble salt of the metal of group VI and optionally formamide;

c) the alcohol dispersion and the aqueous solution are mixed, obtaining a gel;

d) aging of the gel at a temperature ranging from 10 to 40°C;

e) drying of the gel;

f) calcination of the gel.

37. A catalyst according to claim 36, wherein said catalyst has a surface area greater than 300 m²/g.

38 ~~35~~) A catalyst which comprise a metal of group VIII, a metal of group VI, a metal oxide as carrier, a zeolite of the FER type, in a quantity ranging from 5 to 30% by weight with respect to the total weight of the catalyst, and phosphorous in a quantity ranging from 0.1 to 10%.

38 ~~37~~) The catalyst according to claim ~~35~~ ³⁸ containing phosphorous in a quantity ranging from 1 to 5 % wt with respect to the total weight of the catalyst.

40 ~~34~~) A process for preparing the catalyst of claim ~~33~~³⁸
comprising the following steps:

5 a) an alcohol dispersion is prepared, containing a
soluble salt of the metal of group VIII, the zeolite of
FER type and an organic source of aluminum;

b) an aqueous solution is prepared, containing a
soluble salt of the metal of group VI, H_3PO_4 and
optionally formamide;

10 c) the alcohol dispersion and the aqueous solution are
mixed, obtaining a gel;

d) aging of the gel at a temperature ranging from 10 to
40°C;

e) drying of the gel;

f) calcination of the gel.

15 4) ~~35~~) A process for preparing the catalyst of claim ~~33~~³⁸
comprising the following steps:

a) an alcohol dispersion is prepared, containing a
soluble salt of the metal of group VIII, a zeolite of
FER type and an organic source of aluminum;

20 b) an aqueous solution is prepared, containing a
soluble salt of the metal of group VI, and optionally
formamide;

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c) the alcohol dispersion and the aqueous solution are mixed, obtaining a gel;

d) aging of the gel at a temperature ranging from 10 to 40°C;

5 e) drying of the gel; .

f) calcination of the gel.

g) impregnation of the catalyst obtained from step f) with an aqueous solution of H_3PO_4 , drying and calcination.

10 ~~42~~³⁶) Process for preparing the catalyst of claim ~~33~~³⁸ comprising:

a) impregnation of metal oxide with an aqueous solution of H_3PO_4 , drying and calcination,

15 b) impregnation of the material resulting from step a) with an aqueous solution of metal of group VIII and an aqueous solution of metal of group VI,

c) drying and calcination of the material resulting from step b)

20 d) mixing the impregnated metal oxide obtained from step c) with the zeolite of FER type.

43 ~~38~~) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by

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impregnation of the metal oxide carrier with an aqueous solution of H_3PO_4 followed by impregnation with an aqueous solution of the metal of group VIII and an aqueous solution of the metal of group VI.

5 ~~44~~ 39) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by drying and calcination of a gel obtained mixing an alcohol
10 dispersion containing a soluble salt of the metal of group VIII and an organic source of aluminum with an aqueous solution containing a soluble salt of the metal of group VI and H_3PO_4 .

15 ~~45~~ 40) A catalyst which comprises a metal of group VIII, a metal of group VI, a metal oxide as carrier and P in a quantity ranging from 0.1 to 10 % weight with respect to the total weight of the catalyst, prepared by
20 impregnation with an aqueous solution of H_3PO_4 of a gel, dried and calcined, obtained mixing an alcohol dispersion containing a soluble salt of the metal of group VIII and an organic source of aluminum with an

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aqueous solution containing a soluble salt of the metal of group VI.

4641) The catalyst according to claim ⁴³38, ⁴⁴39 or ⁴⁵40 containing phosphorous in a quantity ranging from 1 to 5 % wt with respect to the total weight of the catalyst.

~~42) The process according to claim 1 or 2 wherein the hydrocarbons boiling within the range of 35 to 250°C contain more than 1000 ppm of S.~~

10 43) The process according to claim 1 or 2 wherein hydrocarbons boiling within the range of 35 to 250°C derive from cracking processes.

44) The process according to claim 1 or 2 wherein the catalyst is in extruded form.

15 45) The process according to claim 31 carried out at a ~~temperature ranging from 220 to 330°C.~~